

Sea-salt aerosol in Hawaii and the Influence of Large Organized Structures (LOS) or Rolls on Fluxes and Visibility

Antony D. Clarke

Department of Oceanography

University of Hawaii

1000 Pope Rd., Honolulu, HI 96822

phone: (808) 956-6215 fax: (808) 956-7112 email: tclarke@soest.hawaii.edu

Steven G. Howell

phone: (808) 956-5185 fax: (808) 956-7112 email: showell@soest.hawaii.edu

Vladimir N. Kapustin

phone: (808) 956-7777 fax: (808) 956-7112 email: kpustin@soest.hawaii.edu

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LONG-TERM GOAL

Our long-term goal is to establish an improved understanding of the factors that control the marine aerosol properties and concentrations as they relate to generation processes, mixing processes, their dependence on oceanic and environmental conditions and physicochemical evolution in the marine boundary layer. We expect these efforts to lead to improved modeling and predictability of marine aerosol concentrations and optical properties.

OBJECTIVES

One of the components of our work is to describe the chemical composition of submicron, non-refractory, aerosol over the central Pacific Ocean, a region where little data of this type exists. Our studies involve using an Aerodyne Time-of-Flight Aerosol Mass Spectrometer (ToF-AMS) to chemically characterize the aerosol. We also seek to address the evolving issue of organic aerosol in the marine environment, and deduce potential sources of these organics. More specifically, we wish to continue our investigation of the marine source of organics to the remote marine atmosphere, and add strength to our hypothesis that little to negligible organics can be associated with a natural marine source. Our activities this past year also include analysis of equatorial Pacific aerosol aircraft data (NCAR C-130 PASE experiment) as part of our studying Large Organized Structures (LOS) or rolls and their role in aerosol processes and the vertical mixing of sea-salt and associated optical extinction.

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APPROACH

We have continued analysis of data from the KA-09-03 (TAO) cruise that took place August 24 to September 24, 2009 on board the R/V *Ka'imimoana*. The HR-ToF-AMS was obtained through an ONR supported DURIP proposal for marine aerosol studies and was deployed alongside a nephelometer, APS, LDMA, and a cloud condensation nucleus counter (CCN). Since the cruise ended last year, the focus of the analysis has been on the data from the AMS, which measures non-refractory aerosol masses of organic material (Org) and non-seasalt derived sulfate (nssSO₄). Low concentrations of nssSO₄ and Org over the Central Pacific Ocean suggest a relatively “clean” marine atmosphere in this region, with small continental and anthropogenic influence. However, Org enhancements were encountered on the easternmost leg of the cruise, and potential sources investigated using a combination of air mass back trajectories (AMBT), SeaWIFS, and high resolution AMS data analysis. This analysis has been combined with similar analysis for our VOCALS data off the coast of Chile in 2008 where additional instrumentation quantified combustion sources.

WORK COMPLETED THIS YEAR

The TAO cruise data is being combined with results from our VOCALS study over the Southeast Pacific in a paper titled “Organic carbon and non-refractory aerosol over the remote South Pacific: oceanic and combustion sources”, L. M. Shank, A.D. Clarke, S. Howell, S. Freitag, V. Brekhovskikh, V. Kapustin, C. McNaughton, T. Campos, which is slated for submission in November 2010. This work was also presented at the international Commission on Atmospheric Chemistry and Global Pollution, part of the International Global Atmospheric Chemistry program (iCACGP-IGAC), as a poster entitled “Submicron aerosol over the central Pacific: observations of non-refractory composition under remote “clean” marine conditions”.

In order to investigate potential sources of Org aerosol to this remote region of the ocean, the relationship between Org, Black carbon (BC) and nssSO₄ was examined. Excursions from a cruise-average Org/nssSO₄ ratio of 0.05 are pronounced along the easternmost leg of the cruise, along 125°W from 8°S to 8°N (see Figure 1 for cruise track), where absolute Org concentrations increase gradually from 0.06 at the southern end of the cruise track, to 0.17 µg m⁻³ near the equator, and Org/nssSO₄ values rose from 0.05 to 0.3. This suggests either an additional source of Org to the area, or one with different Org/nssSO₄ ratio. As the ship moved further north (26 Sept), through the ITCZ, the Org/nssSO₄ ratio decreased to values below 0.1, and the absolute value of Org decreased back to concentrations typical of the cruise average 0.06 µg m⁻³.

In VOCALS a linear relationship between Org and BC was observed in the marine boundary layer (Figure 2). Since there is no marine source of BC, this relationship suggests that most, if not all Org in this region is combustion derived. AMS nssSO₄, on the other hand, does not show this same relationship with Org and BC, and clearly shows that when BC goes to zero, there is residual SO₄, part of which may represent marine-source SO₄.

In order to explore the potential source of the Org enhancement during the TAO cruise, two sets of fifteen-day (360-hour), isentropic AMBTs were performed, one on the easternmost leg of the cruise, during the peak of the Org/nssSO₄ excursion, and one on the western edge of the cruise track, where Org/nssSO₄ is close to the cruise-average 0.05 value.

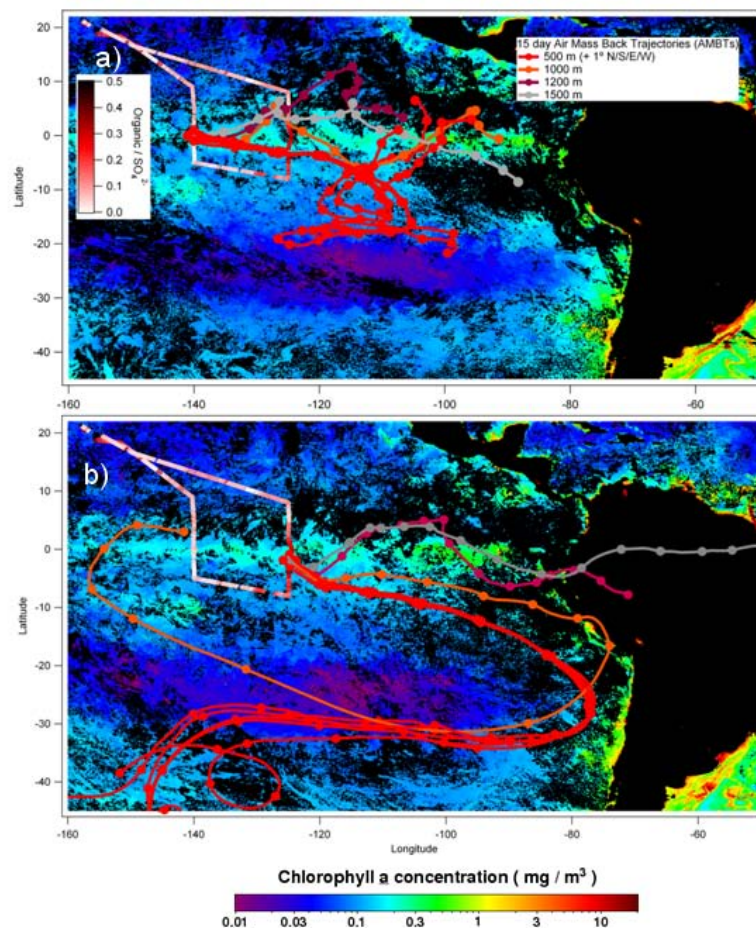


Figure 1. SeaWiFS chlorophyll-a 8 day composite for periods a) 29 Aug-5 Sep 2009 overlaid by AMBTs from 3 Sep 2009 and b) 6 Sep – 13 Sep 2009 overlaid by trajectories from 13 Sep 2009. Cruise track is shown, colored by Org/nssSO4.

Trajectories from 13 September 2009, which is the peak in the Org/nssSO4 ratio, indicate that air masses have had possible continental influence in the past 15 days. Biomass burning in South America serves as a potential source of organics to the FT, and data from the Fire Locating and Modeling of Burning Emissions (FLAMBE) indicates that there were fires in the Amazon 1-2 weeks before sampling occurred on 13 Sept. Eight-day composites of chlorophyll-a concentration were produced using SeaWiFS, to investigate potential biological influences on Org concentrations. However, these do not indicate a significant increase in biological production for trajectories encountered on the western boundary of the cruise track (Figure 1a), versus the eastern edge of the cruise track (Figure 1b).

Further evidence for biomass burning was provided using the AMS high-resolution analysis package. A peak at m/z 60 can be attributed to $C_2H_4O_2$, a fragment generated from the breakdown of levoglucosan, a chemical indicator of biomass burning. Figure 3 shows the Org/nssSO4 ratio time series during the cruise, overlaid by $C_2H_4O_2$ /total Org. There appears to be a relationship between the

Org/nssSO₄ and the C₂H₄O₂/Org ratios, suggesting that perhaps the excursions from the cruise-average Org/nssSO₄ is due to long range transport of biomass burning from the Amazon.

RESULTS

The median concentration of nssSO₄ over the entire cruise was found to be 0.71 $\mu\text{g m}^{-3}$, while Org were 0.06 $\mu\text{g m}^{-3}$ and ammonium (NH₄) median concentrations were 0.07 $\mu\text{g m}^{-3}$. There was essentially no detectable nitrate (NO₃) in this region. In particular, these results indicate small Org contribution, both relative and absolute, to total aerosol mass. Org aerosol enhancement is observed in some instances, and at this time cannot be linked to either a natural or anthropogenic source. Analysis of the combination of AMBTs and satellite derived chlorophyll-a do not lend insight into whether a natural marine source is responsible for the enhancement. However, an increase in a biomass burning indicator (C₂H₄O₂), a product of the breakdown of levoglucosan, suggests a possibility that the enhancement is linked to the long range transport of biomass burning.

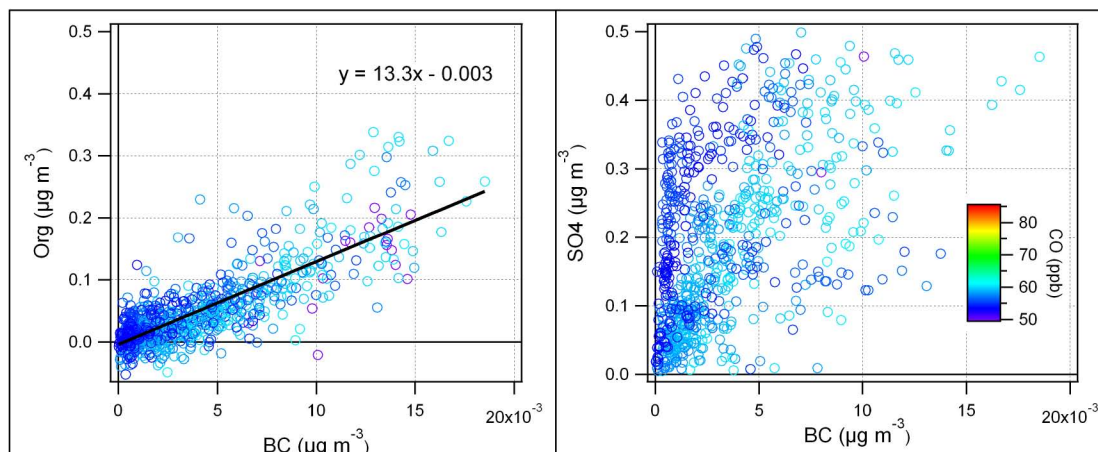


Figure 2. Org vs. BC (left) and nssSO₄ vs. BC (right) in the marine boundary layer during VOCALS.

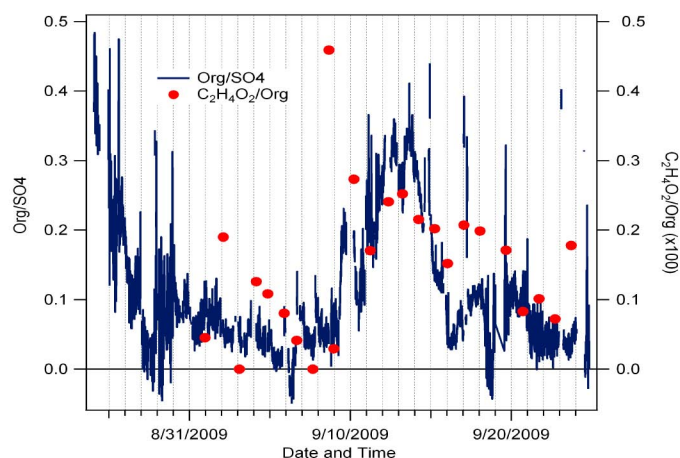


Figure 3. Time series of Org/nssSO₄ and C₂H₄O₂/Org for KA-09-03 cruise

We have also finalized the analysis of LOS influence on aerosol evolution in the MBL and prepared related paper for submission. Roll structures in the diverse MBL settings demonstrate that these can play an active role in the redistribution of aerosol, gas and water vapor in the MBL compared to non-roll cases and lead to changes in the MBL optical parameters relative to regions not influenced by the rolls.

RELATED PROJECTS

No related projects